Supplementary Material:

CONTRIBUTION OF TRANSIENT ELECTRIC FIELDS

The laser excitation may cause some charge redistribution leading to a field which may affect the movement of diffraction with time. To assess the contribution of this transient electric field on the observed diffraction patterns we recorded both the movement of the transmitted direct electron beam and the change in the center of the (110) ring, its integrated intensity and its width, as shown in Fig. 1. At the fluence used (10 mJ/cm^2) a small transient electric field is visible in the movement of the direct beam (at most 0.03 degree) as depicted in Fig. 1(c) (blue curve), with the center of the diffraction ring changing similarly (red curve). Also the change of the direct beam width and the width of the diffraction ring (Fig. 1(b)) have similar magnitude and temporal behavior. Thus, they both can be attributed to spatially inhomogeneous, transient electric fields. In contrast, the integrated intensity of the (110) diffraction ring (Fig. 1(a)) clearly displays a decrease after excitation whereas the the intensity of the direct beam remains constant. Therefore, the intensity change of the (110) is due to structural dynamics and is not caused by a transient electric field effect. This is furthermore corroborated by the fact that the intensity of the (110) ring does not begin to recover within the experimental time window, as was observed for the field-induced deflection and broadening. Our reported results in the main manuscript depend solely on the temporal change of the diffracted intensity as in many other studies made in this laboratory (see e.g. [1–3]).

[1] P. Baum, D.-S. Yang, and A. H. Zewail, Science, **318**, 788 (2007).

[2] F. Carbone, D.-S. Yang, E. Giannini, and A. Zewail, Proc. Natl. Acad. Sci., **105**, 20161 (2008).

[3] A. H. Zewail and J. M. Thomas, *4D Electron Microscopy: Imaging in Space and Time* (Imperial College Press, 2010).

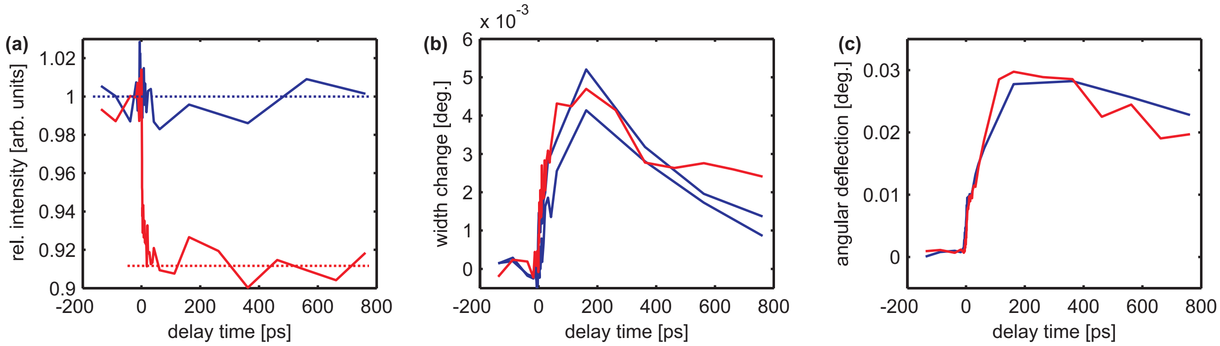

FIG. 1. Comparison between structural dynamics and transient electric field effect. (a)The change of the integrated intensity of the direct beam at different delay times (blue line) is compared to the intensity of the diffracted (110) peak (red line). It can be seen, that, while the direct beam intensity stays constant, the bragg peak intensity decreases due to the Debye-Waller effect and is causes by structural dynamics (b) Due to transient electric fields the width of the direct beam (blue lines for horizontal and vertical width) slightly changes with delay time. The (110) bragg peak shows a comparable broadening (red line), indicating that the broadening is not caused by structural dynamics. (c) The position of the direct beam also shows a small shift vs. delay time (blue line) which leads to a shift in the center of the (110) diffraction ring (red line).